Fiscal Year:	FY 2018	**Task Last Updated:**	FY 05/20/2019
PI Name:	Chaikin, Paul M. Ph.D.		
Project Title:	The Control and Dynamics of Hard Sphere Colloidal Dispersions--NNX13AR67G		
Division Name:	Physical Sciences		
Program/Discipline:			
Program/Discipline--Element/Subdiscipline:	COMPLEX FLUIDS--Complex Fluids		
Joint Agency Name:		**TechPort:**	No
Human Research Program Elements:	None		
Human Research Program Risks:	None		
Space Biology Element:	None		
Space Biology Cross-Element Discipline:	None		
Space Biology Special Category:	None		
PI Email:	chaikin@NYU.edu	**Fax:**	FY 212 995-4016
PI Organization Type:	UNIVERSITY	**Phone:**	212 998-7694
Organization Name:	New York University		
PI Address 1:	Department of Physics		
PI Address 2:			
PI Web Page:			
City:	New York	**State:**	NY
Zip Code:	10003	**Congressional District:**	8
Comments:	NOTE: PI moved to NYU (from Princeton U) in 2005 per A. Hollingsworth in PI's dept (7/2009). Changed email 3/30/2009 (chaikin@princeton.edu no longer valid).		
Project Type:	FLIGHT	**Solicitation:**	98-HEDS-03
Start Date:	09/06/2013	**End Date:**	09/05/2019
No. of Post Docs:	3	**No. of PhD Degrees:**	4
No. of PhD Candidates:	5	**No. of Master' Degrees:**	
No. of Master's Candidates:	0	**No. of Bachelor's Degrees:**	
No. of Bachelor's Candidates:		**Monitoring Center:**	NASA GRC
Contact Monitor:	McQuillen, J.	**Contact Phone:**	216-433-2876
Contact Email:	john.b.mcquillen@nasa.gov		
Flight Program:	ISS		
Flight Assignment:	NOTE: End date changed to 9/5/2019 per NSSC information (Ed., 10/2/19)		
Key Personnel Changes/Previous PI:			
COI Name (Institution):	Hollingsworth, Andrew Ph.D. (New York University)		
Grant/Contract No.:	NNX13AR67G		
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Performance Goal Text:			

NOTE (Ed., March 2014): Continuation of "The Control and Dynamics of Hard Sphere Colloidal Dispersions--NNX08AK04G", grant # NNX08AK04G with the same Principal Investigator (PI), Dr. Paul Chaikin. Colloid science is entering a new era. Over the past 15 years, our NASA-sponsored research has mainly dealt with monodisperse suspensions of colloidal particles interacting via well-known forces. Using spherical particles and observations with light scattering and microscopy, we have gained a great deal of fundamental knowledge about different phases of matter and the dynamics and thermodynamics of their formation. In particular, our experimental results in microgravity have led to a basic understanding of why crystals and glasses form and their properties.

During the past decade, we have made great strides in synthesizing new classes of particles with different shapes and

Task Description:

specific, reversible or irreversible, variable range interactions. We have also found new ways to manipulate the particles with flow, electric and magnetic fields, and light. We are therefore positioned at the threshold of a new technology, assembling equilibrium and non-equilibrium macroscopic structures with function and activity from well designed particles on the nano to micron scale.

Of course, there are still fundamental scientific questions which we can and will address including a host of new ordered phases, frozen configurations, frustration and glasses, and the process of self-organization itself. In particular, we plan to use the microscopy and light scattering instruments, in collaboration with our European colleagues, to study particles that we prepare through emulsion and dispersion polymerization. Physical lithographic techniques will also be employed, and the particles will be modified chemically for controllable interactions. We plan to use different phoretic techniques--electro-, dielectro-, and thermo-phoresis--to control the particles density and orientation. These will also serve as the driving forces to establish the rheological properties of these new systems.

Rationale for HRP Directed Research:

Research Impact/Earth Benefits:

Characterization of crystal formation in the microgravity environment of the ISS (International Space Station) can lead to a greater understanding of how gravity affects many kinds of colloidal materials, including monodisperse ellipsoids and cubes, colloidal clusters of silica or polymer microspheres, DNA-functionalized colloidal spheres, and 'lock-and-key' colloids. By performing these experiments in reduced gravity, we intend to accomplish the desired characterization without gravitationally-induced inhomogeneities that affect both the dynamics and equilibrium state on Earth. Understanding these complex materials should enable new ways of forming ordered phases, such as those sought for photonic devices to be used in optical communication systems. With the ability to make particles of different shapes, i.e., non spherical, we also have the possibility of having directionally dependent particle interactions. For example, we could take tetrahedral clusters of particles and attach DNA to them. The complementary single-stranded DNA 'sticky ends' can associate/dissociate via thermal activation. This arrangement could lead to tetrahedral bonding as found in diamond or in amorphous glass structures. Another approach utilizes depletion interactions. Since we can lithographically prepare particles of any shape we design in two dimensions and many shapes in three dimensions, we can fabricate lock-and-key colloids which only bind to their complementary shape. In this case, the binding is also directional since the congruent surfaces must match. We can also make such lock-and-key particles through emulsion chemistry. Our goal is to produce some simple processes with such ‘designer particles’ and interactions, to lay the foundations for self-assembly and perhaps self-replication of this new class of materials.

Task Progress:

The program’s principle goal is the understanding and explanation of the fundamental microscopic mechanisms of self-organization in model complex fluid systems. The experimental samples are composed of specially synthesized colloidal particles with well understood, well controlled, and sophisticated interactions. Our experiments feature recently developed colloidal systems with directional, specific, and externally controlled inter-particle interactions and motility.
Freezing on a sphere

Using fluorescently-labeled PMMA colloid, we investigated the freezing process of a 2D crystal on a curved surface. The major result is that by employing a novel order parameter, we are able to demonstrate that the topology encodes the position of the disordered regions during the transition, which can be traced to a decreased mobility of the particles. The orientational and icosahedral order change simultaneously and both coincide with the hexatic transition observed in flat-space.

Two dimensional freezing proceeds by the rapid eradication of lattice defects as the temperature is lowered below a critical threshold. But crystals that assemble on closed surfaces are required by topology to have a minimum number of lattice defects, called disclinations that act as conserved topological charges. Consider the 12 pentagons on a classic soccer ball or the 12 pentamers on a viral capsid. Moreover, crystals assembled on curved surfaces can spontaneously develop additional lattice defects to alleviate the stress imposed by the curvature. It is therefore unclear how crystallization can proceed on a sphere, the simplest curved surface on which it is impossible to eliminate such defects.

In our study, we show that freezing on the surface of a sphere proceeds by the formation of a single, encompassing crystalline ‘continent,’ which forces defects into 12 isolated ‘seas’ with the same icosahedral symmetry as footballs and viruses. We use this broken symmetry — aligning the vertices of an icosahedron with the defect seas and unfolding the faces onto a plane — to construct a new order parameter that reveals the underlying long-range orientational order of the lattice. The effects of geometry on crystallization could be taken into account in the design of nanometer- and micrometer-scale structures in which mobile defects are sequestered into self-ordered arrays. Our results may also be relevant in understanding the properties and occurrence of natural icosahedral structures such as viruses.

National Science Foundation (NSF) / Center for the Advancement of Science in Space (CASIS) collaboration

During the period, our project entitled “Nonequilibrium processing of particle suspensions with thermal and electrical field gradients” was recommended for an award. The project team consists of investigators from New Jersey Institute of Technology (NJIT), including Prof. Boris Khusid serving as PI, and the New York University (NYU) team. The main objective of the proposed ISS experiments is to elucidate mechanisms underlying the nonequilibrium assembly of colloidal particles assisted by temperature and electric field gradients and suggest novel routes for processing functional materials. Experiments in the ISS will be conducted to investigate the evolution of phase transitions, instabilities, and the nucleation and growth of crystalline structures in model colloids subjected to temperature and electric field gradients on a fundamental level without the interference of gravity. We hypothesize that the contrast in the structure formation in model colloids under microgravity in the ISS and normal gravity in Earth-based experiments should reveal the salient features of the influence of a temperature gradient, an electric field and gravity on nonequilibrium structure formation in a suspension of interacting particles. These particles are essential for the development and operation of a wide range of terrestrial and space applications. This NSF grant is entirely for complementary support of our NASA supported microgravity studies.

Advanced Colloids Experiment (ACE)-T7: Sample preparation, characterization and operations

An outstanding problem in condensed matter science concerns the relation between particle shape, crystal symmetry, and structure. The simplest and most symmetric crystal is cubic and is naturally comprised of cube-shaped particles. In atomic systems, these are cubic lattices of spherical atoms, made anisotropic by their atomic orbitals; in our crystalline structures, the constituent particles are, in fact, colloidal cubes. Our research goal is to produce such colloidal structures, and study the dynamics of crystal nucleation and growth. ACE-T7 will vary the size and concentration of the depletant in several samples with the goal of seeing the effect on 3D crystallization in microgravity. Prof. Sacanna's group (NYU Chemistry) synthesized the silica cubes and prepared the samples, carefully formulating stable particle suspensions and observing 2D crystallization in the lab.

During ISS Increment 55–56, samples 4–6 were investigated. Several experimental runs were conducted over the seven week period (weeks 12–18). These included Module 2: ACE Module S/N 2009 (702); capillary nos. 1 (sample 4), 2 (sample 5), 3 (sample 6). The three samples were homogenized and temperature gradient experiments were performed. Both surface and bulk crystallization were observed in capillaries 1–3. During six weeks of inactivity, the samples continued crystallizing in microgravity. We proposed an additional run to be performed during the August 12–21 period to resume imaging the colloid along the length of the capillaries in module 2. Our expectation is to observe 3D crystals that may have grown over time. We will focus on previously imaged positions to evaluate growth rate and shape evolution.

Initial observation of cubic colloidal crystals: Summary of June 13, 2018 operations

"Fluorescence is observed throughout the entire length of the capillary and particles can be found with the 100X oil objective, indicating that the capillary is well mixed. We set the left side of capillary #2 to 45°C and the right side to 25°C and took Z-stacks at five ROIs along the length of the capillary over time. We are currently seeing if there is a difference along the length of the capillary, i.e., crystals on the cooler side of the capillary and low particle concentration on the hotter side of the capillary. We have also done some image optimization to make visualizing and processing data easier, as some particles contain a lower amount of fluorophore."

Synthesis of particle samples for ACE-T4 and ACE-T11

Colloidal particles of controlled size are promising building blocks for the self-assembly of functional materials. To support the ACE experimental program, the NYU Colloid Synthesis Facility is developing various materials and colloids including fluorescently-labeled poly(N-isopropylacrylamide-co-acrylic acid) microgel particles (ACE-T4), as well as fluorescently-labeled, poly(12-hydroxystearic acid)-stabilized poly(methyl methacrylate) colloid and trimethoxysilyl-terminated PHS-g-PMMA surfactant (ACE-T11). We have synthesized a new reactive dye (HEMA-modified julolidine rhodol), and are also working on synthesizing a reactive Cy3 cyanine fluorophore for copolymerization with methyl methacrylate and methacrylic acid.

Bibliography Type:	Description: (Last Updated: 05/21/2019)
Articles in Peer-reviewed Journals	Guerra RE, Kelleher CP, Hollingsworth AD, Chaikin PM. "Freezing on a sphere." Nature. 2018 Feb 14;554(7692):346-50. (Erratum in: Nature. 2018 Aug;560(7717):E25) https:// ; PubMed PMID: 29446378 , Feb-2018
Awards	Chaikin PM. "2018 Oliver E. Buckley Condensed Matter Physics Prize, For pioneering contributions that opened new directions in the field of soft condensed matter physics through innovative studies of colloids, polymers, and packing. Awarded by American Physical Society at the March 2018 meeting. March 2018." Mar-2018